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14. ABSTRACT

This report results from a contract tasking University of Groningen as follows: Project theme

Three-dimensional (3D) optical photonic crystals, periodic dielectric composite structures that forbid propagation of electromagnetic waves with a wavelength in the visible region in all directions and for any polarization, have recently entered the center stage of the optical sciences. Information and communication technology, applied coloration science, and even the cosmetics and garment industry have demonstrated a rapidly growing interest in the possibility to 'paint' objects with structurally colored materials having precisely known spectral properties [1-5]. Technical production of photonic crystals is still in its infancy, but clear demonstrations of the feasibility have recently been reported, indicating a rapidly opening research field [4].

Nature has been experimenting already for millions of years with photonic crystals, although this has been recognized only in the last decade. The most striking examples are found among insects and birds, where brilliant, metallic colorations, produced by purely dielectric materials are widely encountered [1-3]. Famous examples are the metallic-green buprestid and golden scarab beetles. The optical origin of these beautiful reflections is well understood, also owing to our recent contributions [3-5]. The beetles feature stacks of multilayers with thickness of the order of 100 nm that act as interference reflectors [5]. We have recently described similar structures in damselflies [5, 6]. Distinctly more complicated are the structural colors of shiny butterflies, e.g. the intense blue Morphos and the ultraviolet reflecting pierids. We nevertheless have gained a reasonable understanding also of this case. Virtually unexplored are the reflections of many butterfly and beetle species where three-dimensional architectures create highly sophisticated photonic crystals [3-4].

Supported by an EOARD grant (March 2006 – March 2007; see project report) we have investigated the 3D-cuticular microstructure in the wing scales of a variety of butterfly species, and we have found that electron microscopical results of the anatomy can be modeled by a gyroid structure [7]. In the species investigated so far, the cuticle volume fraction varies between 0.17 and 0.40 and the lattice parameter is estimated to vary between 165 and 360 nm [7]. Extensive analyses of self-assembled bicontinuous cubic structures demonstrated that the gyroid structure has potential as a 3D photonic crystal [8]. Computer simulations have shown that for relatively large dielectric contrasts (n/n' ≈ 3.5, where n and n' denote the refractive indices of the two composites) the gyroid structure has a complete photonic band gap (PBG) for filling factors between approximately 0.04 and 0.55 [8].

The cuticle/air structures of butterfly wing scales have a refractive index contrast $n/n' = 1.55 \pm 0.05$) + i(0.06 ± 0.01) [9]. Hence, given that for various butterfly species these structures can be modeled by a gyroid structure with a cuticle volume fraction varying between 0.17 and 0.40, these structures are biological photonic crystals without a complete PBG. Yet, partial band gaps may exist for light propagation in some directions or for some polarizations. Because of the absence of complete PBGs, it might be expected that the reflected light from the gyroid cuticular structures is strongly angle-dependent. Experimental proof is so far lacking, presumably because this effect is masked in real

butterfly scales by the fact that one scale is constructed from several grains with different orientations. We are presently studying this aspect further into more detail.

Execution of the project

In this project we will firstly focus on the optical properties of the wing scales of butterflies that have three-dimensional (3D) structures, acting as photonic crystals. The scales of numerous butterflies, specifically several Papilionidae and Lycaenidae, are very likely structured in the form of 3D photonic crystals to achieve a vivid coloration of the wings. We will study the structural details with a combination of scanning and transmission electron microscopy (SEM and TEM), to quantitatively determine the 3D structure of the wing scales. SEM will be performed in close collaboration with the research group of Prof. J.Th.M. De Hosson of the Department of Applied Physics, Materials Science Center, University of Groningen. TEM will be done with the Electron microscope unit of the University of Groningen, and also in collaboration with Prof. T. Hariyama, University of Hamamatsu, Japan.

Angle-dependent reflection and transmission microspectrophotometry data will be collected from isolated, single scales. The results will be interpreted in terms of the photonic band structure of the complex 3D structures. The butterfly wing-scales are expected to be extremely valuable for analyzing angle and wavelength-dependent light scattering. Although we have ample instruments for spectral measurements, we are presently building a novel imaging scatterometer, which will allow us to quickly characterize the optics of butterfly scales with unprecedented detail. We expect to have a working prototype in 3-4 months.

For the polarization studies, collaboration with Dr Dennis Goldstein, who has created in Eglin AFB a unique laboratory for polarization optics, will be of great benefit [10]. The extension of measurements to the infrared will further allow the study of photonic aspects in the thermal wavelength range.

In addition to the experimental measurements, the project will have a large theoretical and computational component. For that we will largely rely on Dr K. Michielsen (consultancy firm EMBD), who has developed very efficient algorithms for calculating the light flux through materials with nanosized dimensions. The computational work will be executed in an intimate relationship with the experiments. Preliminary results have already yielded encouraging results.

Parallel to the work on butterfly wing scales, we will study beetle species that exhibit intense colorations. Scarab beetles are of special interest, because of their circular dichroic reflections. Together with Dr Hariyama we have already made a good inroad into the reflectance properties of buprestid beetles [5, and in preparation] and their function in beetle behavior, and we expect that the collaboration with Dr Goldstein will establish important new insights into the polarization properties of beetles [10] and their role in beetle vision [5].

Explorations of other animal colorations may well reveal unknown coloration techniques. Notably birds and marine animals may be potential gold mines [1, 11]. The proposal to pursue a detailed study of insect (and more general of animal) coloration will not only further the physical field of photonic crystals, but it will also directly stimulate our understanding of insect vision, optical signaling, and animal communication, specifically concerning the aspects of color and polarization. We will continue to study these themes in an intense collaboration with the laboratory of Dr. K. Arikawa, SOKENDAI, Hayama, Japan.

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Detailed work breakdown schedule

We will focus on three specific topics:

- 1. Spectral reflectance of papilionid butterfly wing scales
- July 2007 March 2008: Optical measurements of the scattering of papilionid wing scales as a function of angle and wavelength
- September 2007 March 2008: Electron microscopy of papilionid scales
- July 2007 March 2008: Computational studies to model the optical data using anatomical data

Results: Four papers on optical, anatomical, and computational work (November 2008)

- 2. Spectral reflectance of lycaenid butterfly wing scales
- January 2008 January 2009: Optical measurements of the scattering of several lycaenids wing scales as a function of angle and wavelength
- March 2008 December 2008: Electron microscopy of lycaenid scales
- November 2008 March 2009: Computational studies to model the optical data.

Results: Four papers on optical, anatomical, and computational work (September 2009)

- 3. Optical properties of beetle cuticle and elytra
- April 2008: First characterization of a wide variety of beetles using museum collections by (UV)photography and (micro)spectrophotometry, leading to the identification of key species that deserve detailed study
- August 2008 March 2009 Anatomical studies of beetle cuticle
- December 2008 March 2009: Angle and polarization dependent spectral measurements on key specimens
- May 2008 and February 2009: Optical measurements with Dr. D. Goldstein (in Eglin)
- May 2009 December 2009: Theoretical analysis and computational modeling of optical measurements using the anatomical data

Results: Four papers on optical, anatomical, and computational work (April 2010)

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[FA8655-08-1-3012] – Photonic crystals on the wing Fourth report (1 November 2009 – 30 April 2009)

Summary

The structural colors on the wings of insects, specifically butterflies and beetles, and birds are studied with a variety of optical methods, both experimental and theoretical/computational. In the past half year we have further developed our methods to reveal the intricate physical and photonic properties of butterfly scales, beetle elytra and bird feathers. We furthermore have extended our research to iridescent structures in flowers. The apparent tuning of the coloration of animals and flowers to the spectral properties of visual systems stimulates the investigation of the physical and chemical mechanisms responsible for modifying the reflectance spectra of living systems.

Introduction

Many biological systems are known to use structural colour effects to generate aspects of their appearance and visibility. The study of these phenomena has informed an eclectic group of fields ranging, for example, from evolutionary processes in behavioural biology to micro-optical devices in technological engineered systems. However, biological photonic systems are invariably structurally and often compositionally more elaborate than most synthetically fabricated photonic systems. For this reason an appropriate gamut of physical methods and investigative techniques must be applied so that the systems' photonic behaviour may be appropriately understood. The results, invariably collected using a range of microscopic, optical interrogation and modelling techniques, have been wide-reaching. They have offered insight into a variety of biological topics such as: species' behaviour and communication; crypsis strategies and pressure-driven adaptation methods; and morphological, developmental and evolutionary processes.

Particular recent advances have been made in the study and understanding of animals which exhibit structural color. Structural color comprises the optical effects produced when incident electromagnetic radiation (such as visible, ultraviolet (UV) and near-infrared (NIR) light) encounters ordered spatial variations in a sample's constituent

dielectric material that are on the same length scale as the wavelength of the incident light. A dielectric material is characterized by its refractive index; its value only has a real component when it comprises no optical absorption, and it has both real and imaginary components when light is absorbed. The spatial distribution of the sample's constituent dielectric material, and hence its refractive index, will largely determine the flow of light through or from it. It is this light scattering, in reflection and transmission, which subsequently determines the object's visual appearance.

It is now well-known that, generally, biological objects with inhomogeneous irregular refractive index distributions are colorless or diffusely white when there is no absorption. However, when their constituent refractive index distributions have ordered or quasi-ordered sub-micrometer domains, then wavelength-dependent coherent scattering can occur. This can give rise to bright structural colors. A striking example is one of the icons of structural coloration, the *Morpho* butterfly family. In *M. didius* for instance, the dorsal (upper) wing surfaces display a brilliant blue reflection when illuminated from directly overhead. This color effect vanishes upon oblique illumination (see the figure; from Vukusic and Stavenga 2009).



The upper wings of the tropical butterfly *Morpho didius* have a striking blue color (a) due to multilayer interference reflection. Upon oblique illumination (b), outside the angle of interference reflection, a light brown pigmentary color in the wing scales becomes visible. Note the appearance of "eye-spots" from the wings' ventral surfaces, which are often quite marked in the dorsal wings of other butterflies. With wide-angle illumination the eyespots are completely unnoticed due to the high intensity of the blue interference reflection. (c) TEM image of a cross section through the bilayer of dorsal wing scales on M. didius, showing the highly reflective scale present basally, which is covered by the highly diffractive scale present superficially. Scale bars: (a, b) 1 cm, (c) 1.5 μ m.

Phenomena such as this are controlled by the scales that imbricate *Morpho* wings, as occurs in virtually all Lepidoptera. We study the photonics of the structural colors not only in butterflies, but also in other insects, as well as in birds, and recently also in flowers. Our aim in the longer term is to turn our attention to biomimetic applications.

Methods

We apply mostly experimental optical methods, specifically spectrophotometry and scatterometry, which are complemented by theoretical and computational approaches. We have been recently asked to write a critical review (Vukusic and Stavenga 2009) of the methods used in our research field, which has been published in a beautiful, special issue of the Journal of the Royal Society Interface devoted to various studies of iridescence in nature. The issue includes also a few of our research papers, resulting from the work enabled by the financial support provided by this AFOSR/EOARD grant (http://rsif.royalsocietypublishing.org/site/misc/iridescence.xhtml).

The newly built imaging scatterometer (ISM; described in Stavenga et al. 2009) proves to be a most powerful instrument for unraveling the spatial characteristics of light scattering by natural objects. Our microspectrophotometer (MSP) appears indispensable for the spectral measurements. A further useful setup, consisting of two independently rotatable optical fibers, allows rapid assessment of the angle-dependent reflectance measurement (ARM), including polarization.

Results

We have completed a study on the structural coloration of the Green Hairstreak, *Callophrys rubi*. Extensive experimental measurements have been complemented with three-dimensional finite-difference time-domain (FDTD) calculations for light scattering from differently oriented gyroid structures with model parameters as obtained before (Michielsen and Stavenga, JRSI 2008). A paper describing the new findings has been submitted for publication.





The Green Hairstreak butterfly has ventrally scales consisting of 3D gyroid domains acting as photonic crystals, which can be easily visualized in a polarization microscope (with polarizer P and analyzer A crossed). The domains reflect predominantly yellow green and blue light, into various spatial directions, causing together the matte green appearance of the wings

Two students from Exeter have visited Groningen in January for a week, and three students from Exeter, together with their supervisor, P. Vukusic, have visited Groningen in April for another week, to exploit the optical equipment available, specifically the scatterometer and microspectrophotometer. The brilliant colors of a number of beetle species have been characterized, as well as the wing colors of some papilionid butterflies.

Further work in progress is a collaborative effort with colleagues from Arizona State University on the Pipevine Swallowtail *Battus philenor*. We have identified its blue wing colors to result from combined thin layer and grating interference. A full characterization is underway.

Together with a master student from the Plant Physiology Department of Groningen University we have investigated structural colors of various flowers as part of a larger study of flower colors and insect visitors. Recently a study in Science appeared, claiming that flower iridescence plays a role in pollination. We are presently critically assessing this claim.

A study of the iridescence in Lawes' Parotia, a bird of paradise, together with Dr Daniel Osorio, of the University of Sussex, has yielded exciting insights into the photonics of bird feathers. A paper is in preparation.

Planning of the research program

Our plate is presently loaded with projects that are nearing completion. The next half year will be mostly devoted to finalizing experimental data sets and writing up.

Publications in 2009 so far

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Groningen, 6 May 2009

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